Improved Crystal Quality by Detached Solidification in Microgravity

Liya L. Regel and William R. Wilcox
Clarkson University
Potsdam, NY 13699-5814

E-mail: regel@agent.clarkson.edu

Detached solidification is one of the more surprising, intriguing, misunderstood, and potentially useful observations to come from 25 years of microgravity research. If we can learn how to learn how to reproducibly produce crystals of superior quality by detached solidification, it may be the most important consequence of microgravity research on directional solidification. Such results have sometimes been achieved, but not reproducibly or predictably. This lack of reproducibility stems directly from the fact that the physics underlying detached solidification has not been fully understood. We find ourselves on the threshold of achieving this complete understanding via a new theoretical model.[1-4] This model gives rise to predictions of the conditions required to yield detached solidification. We will test these predictions in collaboration with a recently formed international team. At Clarkson University, we will extend our model to other materials and operating conditions, as well as develop a fully time-dependent simulation. We will attempt to achieve detached solidification on the ground using a transparent, low-melting system that will enable us to see the convection in the melt and the behavior of the freezing interface.

Beginning with Skylab in 1974, many investigators discovered that directional solidification in microgravity often yields ingots that appear to have grown without being in intimate contact with their containers. When this occurred, crystallographic perfection was usually greatly improved -- often by several orders of magnitude. Indeed, under the Soviet microgravity program the major objective was to achieve detached solidification with its resulting improvement in perfection and properties. Although detached solidification has been observed predominantly with semiconductors, it has also been observed with metals and inorganic compounds. This apparent predominance may reflect only the fact that most flight experiments on directional solidification have been performed on semiconductors.

When detached solidification was first discovered, it was generally thought that the melt had lost contact from the ampoule wall because of the high contact angles of the semiconductor melts. This view has persisted, in spite of microgravity experiments [e.g., reference 5] showing that liquids do not pull away from the ampoule wall, no matter what the contact angle. The implicit assumption underlying this model is that the solid took the same shape as the liquid from which it froze. In our model of detached solidification, a meniscus connects the edge of the solid with the ampoule wall, similar to Czochralski growth but with much less distance between the solid and the wall. Because of the curvature of the meniscus and the surface tension of the melt, the pressure in the gap must be greater than that in the adjacent melt. The gas filling this gap consists of one or more volatile constituents that are rejected by the growing solid. In most cases, this is the residual gas remaining in the ampoule that has dissolved in the melt. Although flight ampoules were generally sealed in a vacuum, outgassing from the ampoule wall and the feed material provide adequate gas to fill the gap.

Steady-state numerical simulations using the new model were first performed[2] for InSb, which has exhibited detached solidification in numerous microgravity experiments. These simulations revealed that detached solidification in microgravity is favored by a low freezing rate, increased concentration of volatile constituent, large contact angle of the melt on the ampoule wall (poor wetting), low surface tension of the melt, and a large growth angle. The stability of steady-state detached solidification in microgravity was examined.[3] The shape of the meniscus is destabilizing in a fashion similar to Czochralski growth. If, for example, the crystal begins growing toward the wall, the meniscus shape tends to acceleration the change in diameter. Thus, if only the meniscus is taken into account, one predicts that both Czochralski growth and detached solidification are unstable. Since this is contrary to experimental observations, other factors must stabilize the growth. Dissolved gas transport and heat transfer were examined as potential stabilizing mechanisms for detached solidification. Although gas transport into the gap is usually necessary for detached solidification, it is sufficient to stabilize detachment only for a short distance, on the order of the gap width. On the other hand, heat transfer strongly stabilizes detached solidification, as for Czochralski growth.

Some investigators have chosen to avoid detached solidification by using a spring to press a piston or plug tightly against the end of the melt. Detachment has occurred nonetheless when a plug only lightly contacted the end of the feed ingot. These observations can be explained via the new model by noting that pressing on the melt increases the gas pressure required to maintain the gap between the crystal and the ampoule wall. This is similar to the effect of a gravitational hydrostatic head.[4] In the usual vertical Bridgman configuration, the melt's hydrostatic head must be added to the gas pressure in the gap required to maintain the meniscus shape. Increased transport of gas into the gap is required to maintain this increased pressure. Buoyancy-driven convection can provide this increased transport, provided that the convection is gentle and is directed outward along the freezing interface. On Earth, one would expect such convection for a very slightly convex interface shape. Thus, it is interesting to note that detached solidification was observed on Earth for germanium with a slightly convex interface.[6] Use of a mirror furnace enabled observation of the ampoule in the neighborhood of the freezing interface. The appearance was exactly as expected for our model (i.e., one can see the lines formed by the meniscus where is contacts the wall and the crystal).

References

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